Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

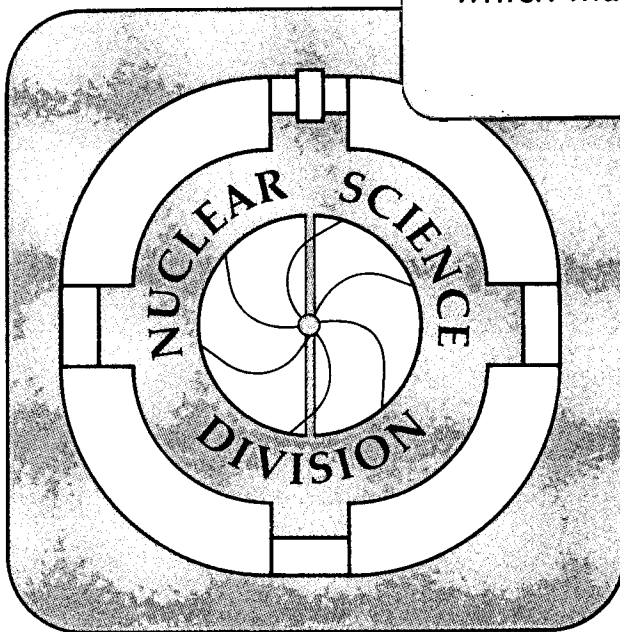Submitted to Nuclear Physics A

Nuclear Penetration Effects in ^{233}U

E. Browne, B. Sur, E.B. Norman, H.L. Hall,
R.A. Henderson, K.T. Lesko, R.M. Larimer,
and D.C. Hoffman

February 1989

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

Nuclear Penetration Effects in ^{233}U

E. Browne, B. Sur, E. B. Norman, H. L. Hall, R. A. Henderson,
K. T. Lesko, R. M. Larimer, and D. C. Hoffman.

Nuclear Science Division
Lawrence Berkeley Laboratory
1 Cyclotron Road
Berkeley, California 94720
USA

Results from precise measurements of the K conversion coefficients for the 300-, 312-, and 340-keV γ rays from ^{233}Pa β^- decay reveal nuclear penetration effects for these predominantly M1 transitions. The experimental conversion coefficients, which are about 18% smaller than values from theory neglecting those effects, remove a previous inconsistency in the transition-intensity balance for the decay scheme of ^{233}Pa, and allow a total 6±2% β^- branch to the ground and first-excited states. The measured value of 8.8±1.4% for this intensity, and the total K x-ray intensity deduced from the present K conversion coefficients, both confirm the revised decay scheme.

RADIOACTIVITY ^{233}Pa [from ^{237}Np α decay]; measured E_γ, I_γ I_{ce}, cc, I_β.

^{233}U deduced nuclear penetration parameter λ.

1. Introduction.

The decay scheme[1] of ^{233}Pa, as shown in Figure 1, consists of three measured β^- transitions that populate the 5/2(633), 3/2(631), and 1/2(631) rotational bands in ^{233}U, respectively, and fifteen γ-ray transitions between levels in these bands. The energies and intensities of the radiations were measured by various authors; γ-ray multipolarities and mixing ratios were deduced from conversion-electron subshell ratios,[2,3,4] and more recently (for the intense 300-, 312-, and 340-keV transitions), from a $\gamma\gamma$ angular correlation measurement.[5]

Gehrke et al.[6] measured 38.6±0.5% for the emission probability of the 312-keV γ ray, and noted that it was inconsistent with the decay-scheme transition intensity balance. Using this emission probability and theoretical conversion coefficients[7] for normalizing the decay scheme, a value of 102±2% can be deduced for the total photon plus conversion electron emission probabilities of the transitions to the ground-state rotational band. This result implied that there is no direct β^- population of this band, and hence is in disagreement with the total measured intensities of 12%[8] or 5%,[9,10] for the β^- transitions to the ground and first-excited states.

Another inconsistency, also noted by Gehrke et al.[6] originates from the total K x-ray intensity as measured in the decay of ^{233}Pa. This intensity, which is about 18% smaller than that deduced from experimental γ-ray data and theoretical conversion coefficients,[7] suggests that the K conversion coefficients for the 300-, 312-, and 340-keV γ rays are smaller than the corresponding theoretical values. The experimental K conversion coefficient of 0.69±0.07 reported for the 312-keV γ ray (Bisgard et al.[8]) is in fact smaller than the theoretical value[7] of 0.78 (M1 multipolarity), but not significantly smaller, considering the large experimental uncertainty.

The two inconsistencies between the measured and decay-scheme deduced

intensities obviously imply smaller conversion coefficients than those from theory.[7] This paper presents precise measurements of the K conversion coefficients for the 300-, 312-, and 340-keV γ rays, and explains these anomalously small results by including nuclear penetration effects in the description of the conversion process. It also presents measurements of the emission probability for the 312-keV γ ray, and of the total β⁻ transition intensities to the ground and first-excited states.

2. Source Preparations.

The ^{233}Pa activity used for this study was chemically separated from ^{237}Np parent activity by means of ion-exchange column techniques. The Np-Pa sample, dissolved in 12 M HCl, was adsorbed onto a Dowex AG1-X8 anion-exchange column. Washes with 12 M HCl removed stable contaminants and fission products. The Pa was eluted off the column with 12 M HCl/0.2 M HF. A 0.5-μCi source, mixed with ^{113}Sn, was prepared by evaporating a solution containing the activities to dryness on a 0.075-mm Ni foil. ^{113}Sn emits an intense 392-keV γ ray with a precisely known K conversion coefficient, and was therefore included for normalizing the γ-ray and conversion-electron intensity scales in the conversion coefficient measurements. A second source, containing only ^{233}Pa, was deposited on a 0.075-mm Ni foil, evaporated to dryness, and then flamed. Its activity of 0.30±0.02 μCi was determined by absolute counting of the 312-keV γ ray with a Ge detector of known absolute photopeak efficiency. This source was used for measuring the β⁻ spectrum of ^{233}Pa with a βγ anticoincidence technique. Finally, two sources for singles γ-ray measurements, one containing 0.01 μCi of ^{233}Pa, and the other, 0.2 μCi of ^{237}Np with ^{233}Pa in equilibrium, were deposited on 0.075-mm Ni foils, flamed, and covered with adhesive tape. No γ rays were observed other than those from ^{233}Pa, ^{237}Np, and ^{113}Sn.

3. Equipment and Calibrations.

3.1 Conversion Coefficient Measurement.

Conversion coefficients for the 300-, 312-, and 340-keV γ-ray transitions were determined from simultaneous measurements of their γ-ray and conversion-electron intensities. γ rays were detected by a high-purity Ge coaxial detector with a volume of 110 cm^3, and conversion electrons, by a 15-mm diameter, 4-mm thick windowless lithium-drifted silicon detector. The detectors faced each other, with the ^{233}Pa-^{113}Sn source mounted in vacuum at 7 mm from the Si(Li) detector, and at 80 mm from the Ge detector. The Si(Li) detector was collimated with a 0.5-mm thick Ta foil to reduce the detection geometry to about 1%, thereby decreasing β⁻-conversion-electron coincidence summing effects. γ-ray and conversion-electron spectra, measured with energy resolutions (full-width at half-maximum) of FWHM=1 (for the 122-keV γ ray from ^{57}Co) and 3 keV, respectively, were accumulated in two 4096-channel analog-to-digital converters. The data were stored on the magnetic disk of an IBM PC/AT computer, and later analyzed with the code SAMPO,[11] using a VAX-8650 computer. The relative photopeak efficiency of the Ge detector was measured as a function of the γ-ray energy with sources of ^{152}Eu and ^{133}Ba, and applied to the 312- and 392-keV γ rays from ^{233}Pa and ^{113}Sn, respectively, through a least-squares interpolation procedure. The conversion-electron energies were calibrated through the most intense lines of well-known transitions from the ^{233}Pa-^{113}Sn source. The conversion-electron intensities were obtained from the spectral areas of the full-energy peaks (same detector response for all electron energies). The value 0.437±0.007 for the K conversion coefficient of the 392-keV transition from ^{113}Sn (Hansen et al.[12]) was used to normalize the γ-ray and conversion-electron intensity scales.

3.2 γ-ray Singles Measurement.

The method used for measuring the emission probability of the 312-keV γ ray from the β⁻ decay of ^{233}Pa was similar to one used by Vaninbroukx et al.[13] It used the known emission probability of 12.3±0.2%[14] for the 86.5-keV γ ray from the decay of ^{237}Np, and the observed ratio between the relative intensities of this γ ray and the 312-keV γ ray in a spectrum from an equilibrium source of ^{237}Np and ^{233}Pa. Because ^{233}Pa emits an 86.6-keV γ ray, the experimental ratio was corrected after determining the same intensity ratio in a spectrum from a separate ^{233}Pa source. The ratio between the detector photopeak efficiencies for 312 and 86.6 keV, which was needed in the data analysis, was determined from the same ^{233}Pa spectrum, using the relative intensities given by Gehrke et al.[6] for these γ rays. The γ-ray spectra were measured with both 110 cm^3 coaxial Ge and 89 cm^3 planar Ge detectors, with sources placed at distances of 230 mm and 300 mm from the detectors, respectively, to reduce possible coincidence summing effects.

3.3 β⁻ Transition Intensity Measurement.

The total intensity of the β⁻ transitions to the ground and first-excited states was measured with a 1-mm thick Au-Si surface-barrier detector, at room temperature, shielded by an annular NaI detector that covered >95% of 4π geometry. The system, operated in anticoincidence mode, significantly reduced the detection of γ rays and β⁻ transitions coincident with γ rays of energies greater than 30 keV, the threshold energy for the NaI detector. The β⁻ transitions to the ground and first-excited states are not in coincidence with such γ rays, except for the very weak (=0.02%) 40-keV γ ray from the first excited state. Their total spectral intensity therefore contained mostly events from those β⁻ transitions, without contribution from other radiations, and thus provided a more reliable value for the intensity that one would obtain from a β⁻ singles

measurement. The β^- spectrum was measured with an efficiency of 2.1±0.3%, low enough to reduce to less than 1% the number of possible events in the region of interest originating from true coincidence summing with conversion electrons. The energy resolution was FWHM=14 keV for the 364-keV K electron line of ^{113}Sn. The energy scale was calibrated with conversion electrons from the intense 312-keV γ-ray transition, and the region from 372 keV to 570 keV was analyzed using a peak shape obtained from the 364-keV K conversion electron line of a pure ^{113}Sn source. The same data acquisition system described in section 3.1 was also used here.

4. Results.

4.1 Conversion coefficients.

A portion of the spectrum of conversion electrons emitted by the ^{233}Pa-^{113}Sn source is shown in Figure 2. The K conversion coefficient for the 312-keV transition was determined to be 0.64±0.02 by measuring the γ-ray and K conversion-electron intensities of the 312-keV and 392-keV transitions from ^{233}Pa and ^{113}Sn, respectively. A correction of 1% for coincidence summing was applied to the conversion-electron intensity of the 312-keV γ-ray transition. This value was estimated from the detection geometry of the experiment. Although the relative γ-ray intensities measured here are in excellent agreement with those reported by Gehrke et al.,[6] the latter had smaller uncertainties and were therefore used for determining the conversion coefficients of the 300- and 340-keV γ rays (see Table 1). The total relative K x-ray intensity of 83±2, as deduced from the present K conversion coefficients, the γ-ray intensities of Gehrke et al.[6] and a fluorescence yield[15] of 0.972, agrees well with the measured intensity[6] of 79±3. Table 1 shows the conversion-electron data for the 300-, 312-, and 340-keV transitions. Values from other authors have been included in this table for comparison.

Subshell ratios, measured here and in earlier[2,3] work, show that the three transitions are predominantly M1. $\gamma\gamma$ angular correlation results of Krane[5] confirm this

conclusion, and mixing ratios (δ), determined in that work, are given in column 6 of Table 2. As the present study shows, however, the experimental conversion coefficients for these transitions are 18% smaller than their theoretical values[7] for M1 multipolarity. Such a discrepancy, which can be explained in terms of nuclear penetration effects in the conversion process,[16] is often significant for M1 γ rays with electromagnetic transition probabilities much smaller than corresponding single-particle Weisskopf[17] estimates. This seems to be the case for the 300-, 312-, and 340-keV γ rays, whose Weisskopf[17] hindrance factors are about 180, 300, and 400, respectively. These values were deduced from the present γ-ray data, and the level half-lives shown in the decay scheme of Figure 1. Nuclear penetration effects are also expected from the Nilsson-Rasmussen[18] selection rules for the asymptotic quantum numbers in deformed nuclei. These rules suggest both photon retardation and allowed penetration in the conversion process for the M1 transitions between the 3/2(631) and 5/2(633) Nilsson orbitals.

The following expression for the conversion coefficients of M1 transitions[15] includes the effect of nuclear penetration

$$\beta = \beta_0 \ (1 + B_1\lambda + B_2\lambda^2) \ , \tag{1}$$

where β_0 is the normal conversion coefficient as tabulated by Rosel et al.[7] B_1 and B_2 are penetration coefficients that have been calculated from atomic-electron wave functions (also tabulated by Hager and Seltzer[19]), and

$$\lambda = \frac{<I' \mid P_1 \mid I>}{<I' \mid M_{1\gamma} \mid I>} \tag{2}$$

is the penetration parameter for a transition between states I and I', respectively. The numerator of equation (2) is the reduced M1 penetration matrix element, and the denominator, the reduced matrix element for γ-ray emission. For example,

$B_1 = -0.080$ and $B_2 = 0.0016$ for K-shell conversion of the 312-keV γ-ray transition. Column 7 of Table 2 contains the nuclear penetration parameters for the three γ rays. These parameters were deduced from the experimental conversion data and equation (1), with the minimization procedure and computer code of Rysavy *et al.*[20] Theoretical values for a transition between the 3/2(631) and 5/2(633) orbitals were calculated from equation (2), taking a typical ^{233}U nuclear deformation of $\delta=0.25$. The results ranged from 3.1, for a gyromagnetic ratio $g_s=g_{s\ free}=-3.287$, to 4.6, for $g_s=0.6\ g_{s\ free}$. The reduced penetration matrix elements used in this calculation were from Krpic *et al.*[21] and the corresponding matrix elements for γ-ray emission, from Browne and Femenia[22] The agreement between the experimental and theoretical nuclear penetration factors confirms the interpretation given above for the anomalous experimental conversion coefficients.

4.2 Emission Probability of the 312-keV γ ray.

The emission probability of the 312-kev γ ray, as deduced from a singles γ-ray spectrum using a ^{237}Np source with ^{233}Pa in equilibrium, is given by

$$\gamma_{312}(\%) = \gamma_{86.5}^{Np}(\%)\times\frac{I_{312}}{I_{86.5}}\ , \tag{3}$$

where I_{312} and $I_{86.5}$ are the relative intensities of the 312- (from ^{233}Pa) and 86.5-keV (from ^{237}Np) γ rays, respectively, and $\gamma_{86.5}^{Np}(\%)$, the emission probability of the 86.5-keV γ ray from ^{237}Np. The ratio $\dfrac{I_{312}}{I_{86.5}}$ can be deduced from the spectral areas of the corresponding γ rays, corrected for the contribution of a weaker 86.6-keV γ ray from ^{233}Pa, and expressed as follows:

$$\frac{I_{312}}{I_{86.5}} = \frac{X^{Pa}}{X^{Np-Pa} - X^{Pa}}\times\frac{1}{Y^{Pa}}\ . \tag{4}$$

X^{Pa} is the ratio between the spectral areas of the 86.6- and 312-keV γ rays for a ^{233}Pa

source, and Y^{Pa}, the ratio between the efficiency corrected intensities of the same γ rays. These ratios are of course not equal because of the differing efficiencies of the Ge detector for the two energies. X^{Np-Pa} is the ratio between the spectral areas of the 86- (86.5 plus 86.6) and 312-keV lines from an equilibrium ^{237}Np-^{233}Pa source. A value of 0.160±0.005 was measured for the first factor given in equation (4), using the spectral areas deduced from two independent measurements with different Ge detectors. Y^{Pa}=0.051±0.003[6] was used to correct the spectral areas of the 312- and 86.6-keV γ rays for the efficiency response of the Ge detector. The intensity ratio $\dfrac{I_{312}}{I_{86.5}}$ then becomes 3.14±0.21, with the precision of the result essentially that of the quantity Y^{Pa}, i.e., 6%. This value agrees well with the more precise ratio of 3.11±0.09 of Vaninbroukx et al.[13] Finally, an emission probability of 38.6±2.6% was obtained for the 312-keV γ ray by using equation (3) and the recommended value[14] of 12.3±0.2% for the 86.5-keV γ ray from ^{237}Np. The central value of this quantity is the same as that reported by Gehrke et al.[6] but its uncertainty is significantly larger.

4.3 β⁻ Intensity to ground and first-excited states.

The β⁻ spectrum of ^{233}Pa measured in this work is shown in Figure 3. Because of the presence of conversion electrons up to about 420 keV, only the region above this energy was used to measure the β⁻ intensity to the ground and first-excited states. The distortions of the spectrum due to electron backscattering in the Au-Si detector were corrected with a data analysis technique of Charoenkwan.[23] Peak shapes were determined from the K electron line of the 392-keV transition from a separate ^{113}Sn source. After the tails due to backscattering were removed, the spectrum of ^{233}Pa was plotted in 14-keV bins. Figure 4 shows the binned spectrum in the region between 380 and 580 keV. A calculated spectrum for a first-forbidden transition, given in the same figure, was fitted to the experimental data points. This procedure gave a value of

8.8±1.4% for the total intensity range of the β^- transitions to the ground and first-excited states. This result is consistent with 6±2%, which can be obtained from the decay-scheme transition intensity balance, using normalized conversion electron intensities from this work and γ-ray intensities from Gehrke et al.[6]

5. Conclusions.

This paper shows that the use of theoretical conversion coefficients for transitions affected by nuclear penetration effects leads to incorrect results, and thereby illustrates the need for precise measurements of these quantities. Both experimental γ-ray conversion coefficients and experimental emission probabilities provide information needed to verify the consistency and correctness of decay-scheme transition intensity balances. Further, this paper shows the usefulness of experimental x-ray intensities for deducing total intensities from conversion for β^- emitters.

References

[1] Table of Isotopes, ed. C. M. Lederer and V. S. Shirley,

(John Wiley and Sons, Inc., New York, 1978) p. 1428

[2] R. G. Albridge, J. M. Hollander, C. J. Gallagher, and J. H. Hamilton, Nucl. Phys.

27 (1961) 529 [3] Georg Schultze and Jurgen Ahlf, Nucl. Phys. 30 (1962) 163

[4] S. A. Woods, P. Christmas, P. Cross, and S. M. Judge, Nucl. Instr. and Meth.

A264 (1988) 333

[5] K. S. Krane, Nucl. Phys. A459 (1986) 1

[6] R. J. Gehrke, R. G. Helmer, and C. W. Reich, Nucl. Sci. and Eng. 70 (1979) 298

[7] F. Rosel, H. J. Fries, K. Alder, and H. C. Pauli, At. Data and Nucl. Data Tables

21 (1978) 291

[8] K. M. Bisgard, P. Dahl, P. Hornshoj, and A. M. Knutsen, Nucl. Phys. 41 (1963) 21

[9] W.D. Brodie, Proc. Phys. Soc. 6 (1954) 397

[10] Ong Ping Hok and P. Kramer, Physica 21 (1955) 676

[11] J. T. Routti and S. G. Prussin, Nucl. Instr. and Meth. 72 (1969) 125

[12] H. H. Hansen, E. De Roost, D. Mouchel, R. Vaninbroukx, Int. J. Appl. Radiat.

Isot.22 (1971) 1

[13] R. Vaninbroukx, G. Bortels, and B. Denecke, Int. J. Appl. Radiat. Isot. 35

35 No. 9 (1984) 905

[14] Decay Data of the Transactinium Nuclides, International Atomic Energy Agency

Technical Reports Series No. 261 (1986) p. 83

[15] M. O. Krause, J. Phys. Chem. Ref. Data 8 (1979) 307

[16] E. L. Church and J. Weneser, Ann. Rev. Nucl. Sci. 10 (1960) 193

[17] J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics,

(John Wiley and Sons, Inc., New York, 1952) p. 627

[18] S. G. Nilsson and J. O. Rasmussen, Nucl. Phys. 5 (1958) 617

[19] R. S. Hager and E. C. Seltzer, Nucl. Data Tables **A6** (1969) 1

[20] M. Rysavy and O. Dragoun, Comp. Phys. Comm. **19** (1980) 93

[21] D. K. Krpic, I. V. Anicin, and R. B. Vukanovic, Nucl. Data Tables **11** (1973) 553

[22] E. Browne and F. R. Femenia, Nucl. Data Tables **10** (1971) 81

[23] P. Charoenkwan, Nucl. Instr. and Meth. **34** (1965) 93

Table 1. Conversion Electron Data for the 300-, 312-, and 340-keV transitions.

$E_\gamma^\dagger$(keV)	Shell	Relative Conversion-Electron Intensities				I_γ^*(rel) Ref. [6]	Conv. Coef.‡ Present work
		Ref. [2]	Ref. [3]	Ref. [4]	Present work		
300	K	193	210	182±4	188±1	17.14±0.15	0.70±0.02
	L_1+L_2	34	35	32±4	34.4±0.3		0.128±0.004
312	K	1000	1000	1000±5	1000±2	100	0.64±0.02
	L_1+L_2	186	164	188±17	192±2		0.123±0.004
	M_1	37	43		46.5±0.6		0.029±0.001
340	K	85	93	93±7	90.4±0.2	11.57±0.10	0.50±0.02
	L_1+L_2	21	17		16.2±0.4		0.090±0.003
	M_1	6	3.3		4.0±0.1		0.022±0.001

† Nominal γ-ray energy.
* Relative γ-ray intensity.
‡ From relative conversion-electron and γ-ray intensities given in columns 6 and 7, respectively, normalized to 0.64±0.02 for the 312-keV γ ray.

Table 2. Conversion Coefficients and Nuclear Penetration Parameters for the 300-,
312, and 340-keV transitions.

$E_\gamma^\dagger$(keV)	Shell.	Conversion Coefficients			Mixing Ratio[*]	Penetration Parameter[‡]
		Experimental	M1[&]	E2[&]	(δ)	(λ)
300	K	0.70±0.02	0.87	0.076	-0.08±0.02	2.6±0.9
	L_1+L_2	0.128±0.004	0.165	0.065		
312	K	0.64±0.02	0.78	0.070	-0.10±0.01	2.3±0.4
	L_1+L_2	0.123±0.004	0.148	0.056		
	M_1	0.029±0.001	0.032	0.0036		
340	K	0.50±0.02	0.61	0.060	-0.23±0.05	2.4±0.8
	L_1+L_2	0.090±0.003	0.116	0.042		
	M_1	0.022±0.001	0.025	0.0030		

† Nominal γ-ray energy.
&Theoretical value from reference [7].
* Recommended value from reference [5].
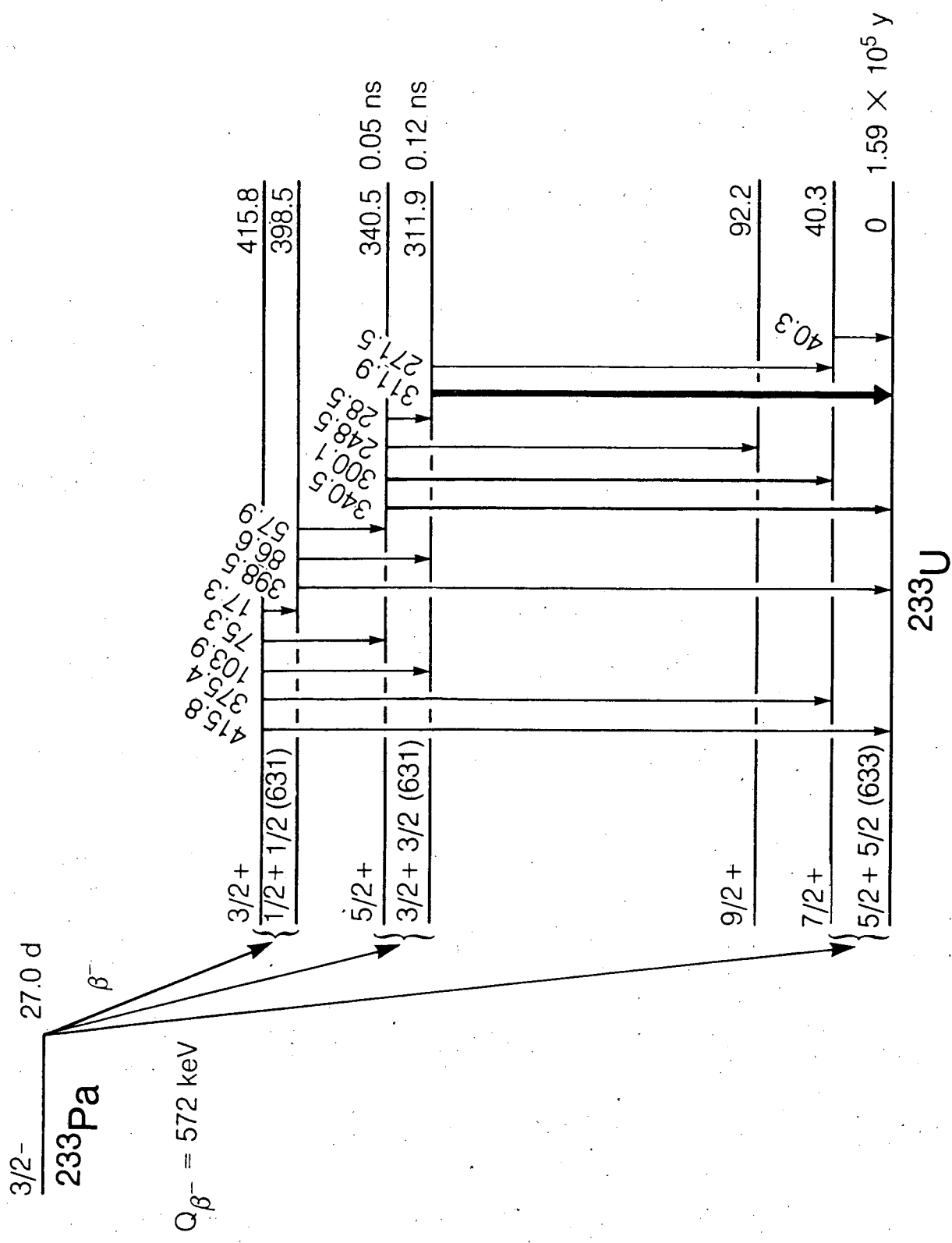‡ Deduced assuming pure M1 multipolarity.

Figure Captions

Figure 1. Simplified [233]Pa decay scheme from reference [1].

Figure 2. A portion of the singles spectrum of conversion electrons emitted by the [233]Pa-[113]Sn source.

Figure 3. [233]Pa spectrum of β^- particles and conversion electrons in anticoincidence with γ rays.

Figure 4. [233]Pa binned spectrum of β^- particles between 380 and 580 keV. The histogram shows the experimental intensity in 14-keV bins. The continuous line correspond to a theoretical shape for first-forbidden transitions to the ground and first-excited states with a best total fitted branching of 8.8%.
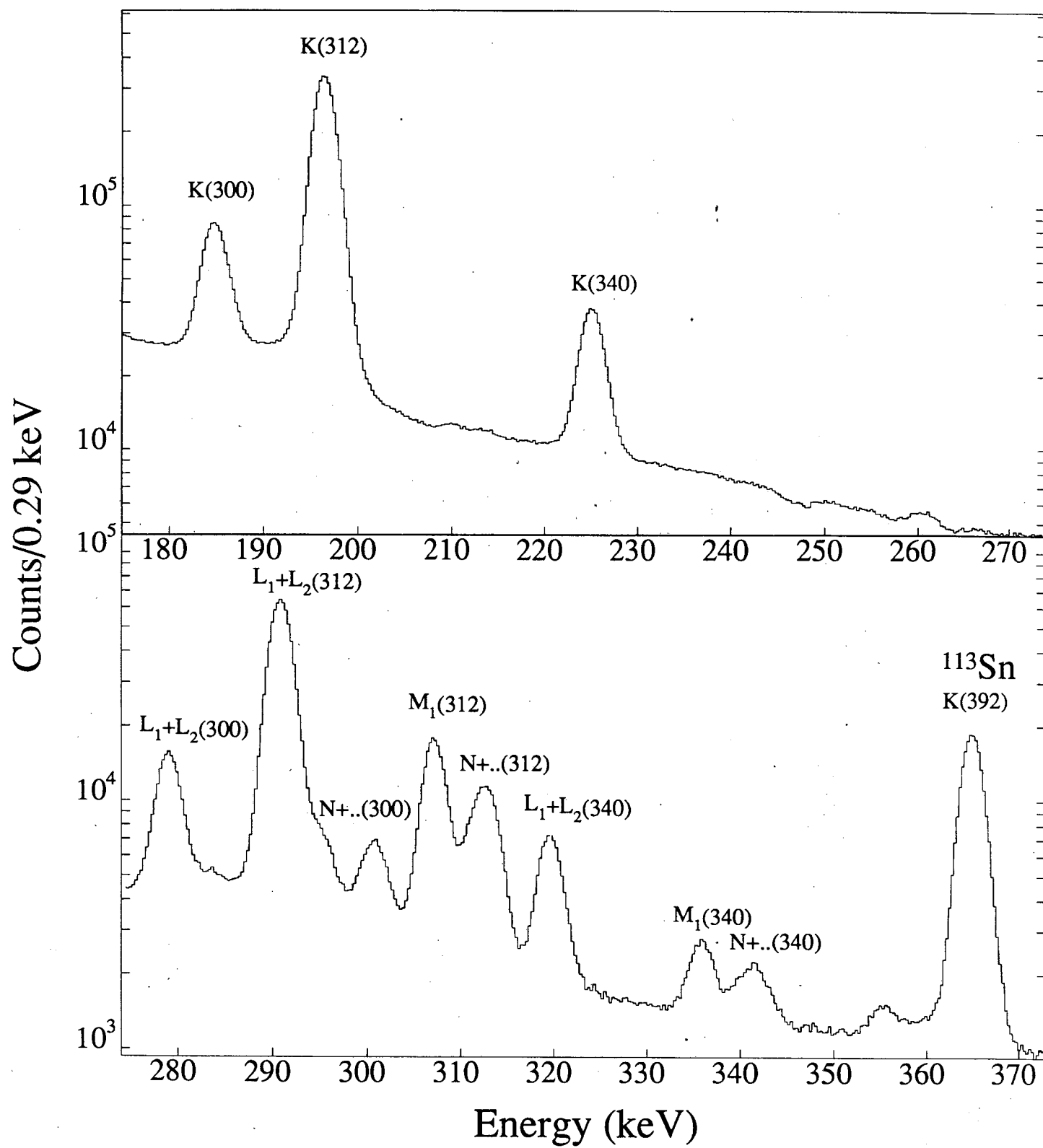
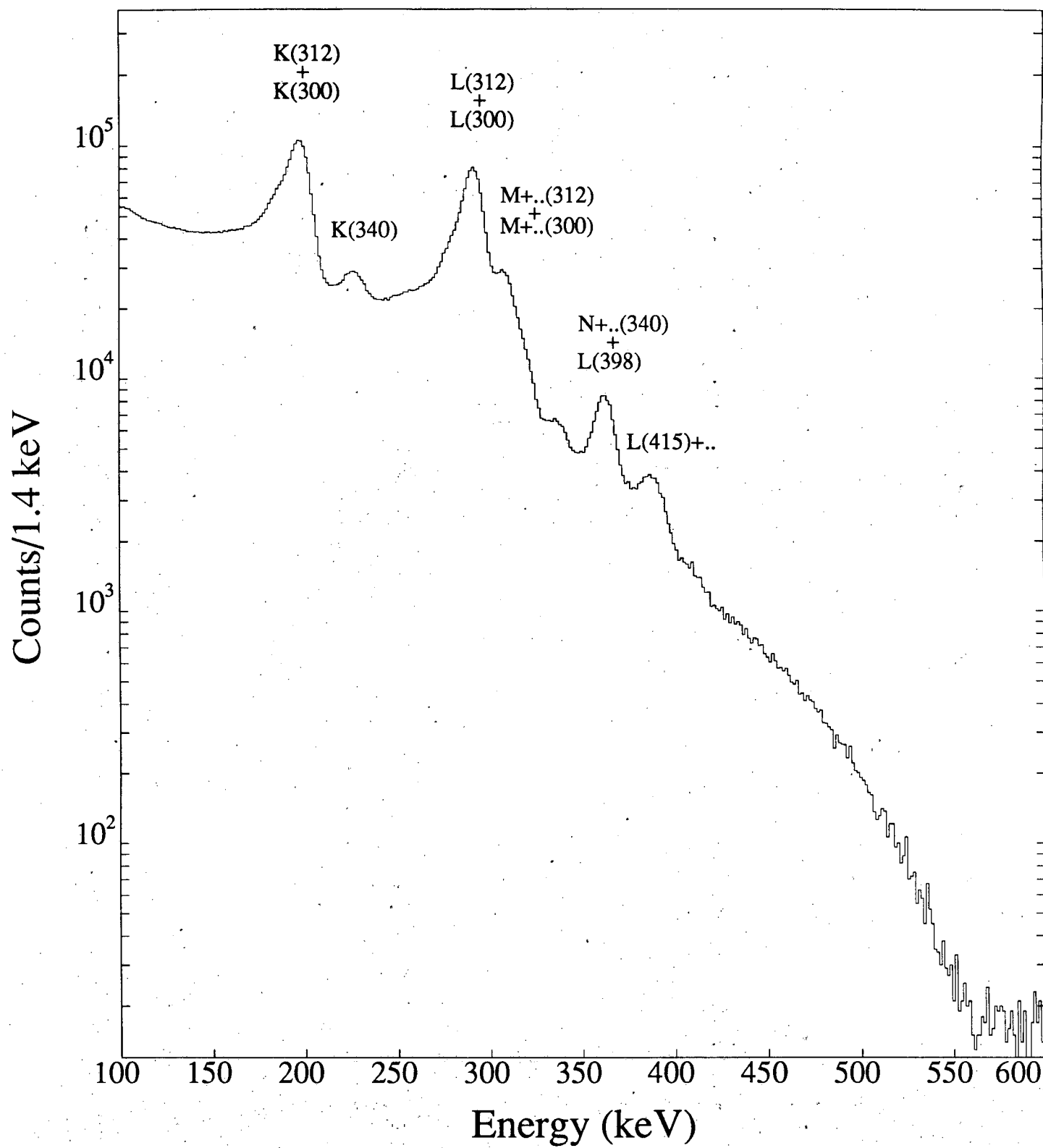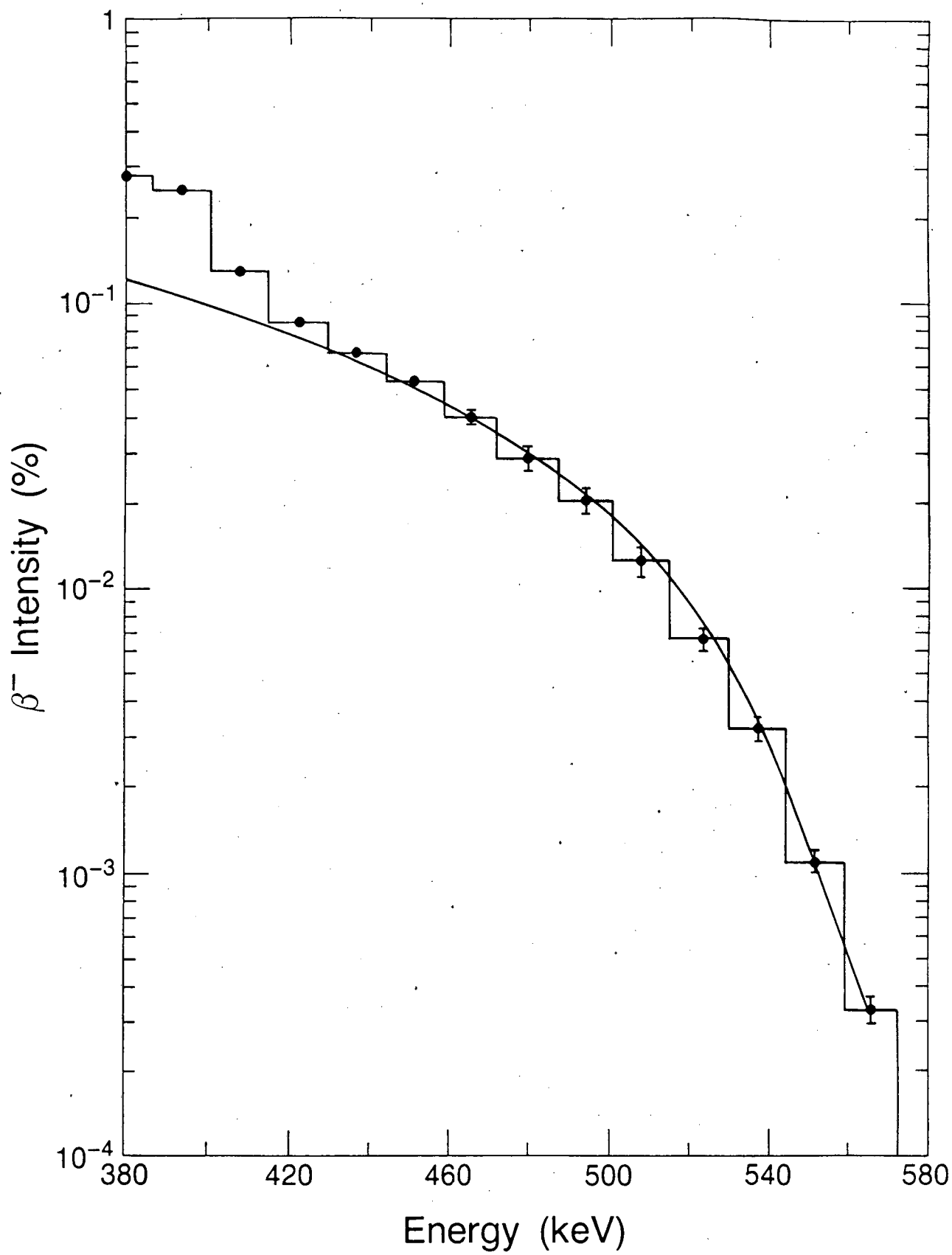Figure 1

XBL 891-7016

Figure 2

Figure 3

Figure 4

XBL 891-7037